

Electron spin decoherence in isotope-enriched silicon

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Silicon is promising for spin-based quantum computation because nuclear spins, a source of magnetic noise, may be eliminated through isotopic enrichment. Long spin decoherence times T_2 have been measured in isotope-enriched silicon but come far short of the $T_2 = 2T_1$ limit. The effect of nuclear spins on T_2 is well established. However, the effect of background electron spins from ever present residual phosphorus impurities in silicon can also produce significant decoherence. We study spin decoherence decay as a function of donor concentration, ^{29}Si concentration, and temperature using cluster expansion techniques specifically adapted to the problem of a sparse dipolarly coupled electron spin bath. Our results agree with the existing experimental spin echo data in Si:P and establish the importance of background dopants as the ultimate decoherence mechanism in isotope-enriched silicon.

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Long electron spin decoherence times in silicon are of significant interest in producing low-error rates for quantum computation. Very long spin echo decay times T_2 have been reported [1–3] in isotope-enriched silicon (i.e., reduced nuclear spin concentration). Resource requirements for quantum error correction are significantly reduced as the qubit fidelity improves, which motivates better understanding of the limits of T_2 even with isotope enrichment. The ultimate decoherence time is theoretically limited by inelastic decay mechanisms (spin-lattice relaxation) on a time scale of T_1 . The increase of T_2 upon reducing the ^{29}Si nuclear spin concentration [1, 2] is now well understood [4, 5]. However, even the highest purity Si wafers contain traces of dopant impurities, usually phosphorus, at levels $\sim 10^{12} - 10^{14} \text{ cm}^{-3}$. Their electron spins are coupled by dipolar interactions, causing fluctuations that induce qubit spin dephasing. In this Letter, we develop the necessary theory to examine decoherence of a central spin in a sparse bath of nuclear and electron spins. We find excellent agreement with existing Si spin echo data showing that existing spin decoherence measurements in Si may already be limited by the coupling of the donor electron spin to the P donor spin bath rather than the Si nuclear spin bath. As a consequence, further isotopic enrichment, an extremely expensive procedure, may not provide any more advantage in the eventual construction of a Si spin quantum computer. In fact, we find that in the presence of donor-induced spin decoherence, T_2 may actually increase when some ^{29}Si is present.

We study here the central spin decoherence problem of a donor electron spin among spins of other donors and ^{29}Si . Because of coupling among the spins, a particular donor electron spin will experience fluctuations of its energy splitting in a phenomenon known as spectral diffusion (SD). ^{29}Si -induced SD calculated using a cluster expansion technique [4], well approximated at the

lowest order with independent contributions from each pair [6], is in excellent agreement with experiments for Si:P [1, 2, 7] and Si:Bi [8] donors. With a firm foundation rooted in a precise quantum mechanical formulation, this was a significant advance over the long history of phenomenological, stochastic models [9–11]. These previous techniques [4, 6], however, are applicable to relatively dense and weakly-coupled spin baths and cannot accurately treat SD due to randomly located donors in which the strength of interaction to the central spin is no different than between bath spins; neither can they handle very low concentrations of ^{29}Si rigorously. A disjoint cluster approach was applied to the relatively sparse bath of carbon spins for the SD of nitrogen-vacancy defects in diamond [12]. Exact numerics were applied [13] in the central spin decoherence problem of dilute dipolarly-coupled spins. Our approach in this Letter is based upon the cluster correlation expansion (CCE) [14] that reformulates the cluster expansion technique [4] such that a large bath approximation is not necessary, making new regimes of the SD problem accessible.

We consider an ensemble of Si:P donor electron spins over varied donor concentrations, C_E (for electron), and ^{29}Si concentrations, C_N (for nuclear). We use parts per million (ppm) of lattice sites for C_N . We include dipolar and hyperfine interactions among spins. Assuming a large applied magnetic field (100 mT is sufficient) in the z direction, we use an effective Hamiltonian in which Zeeman energies are conserved among the electron and nuclear spins independently, allowing only flip-flop dynamics: $\hat{\mathcal{H}} = \hat{H}_E + \hat{H}_N + \hat{H}_{E-N}$, where

$$\hat{H}_E = \sum_{i>j} \gamma_E^2 d(\mathbf{R}_i - \mathbf{R}_j) [\hat{S}_i^+ \hat{S}_j^- + \hat{S}_i^- \hat{S}_j^+ - 4\hat{S}_i^z \hat{S}_j^z], \quad (1)$$

$$\hat{H}_N = \sum_{n>m} \gamma_N^2 d(\mathbf{r}_n - \mathbf{r}_m) [\hat{I}_n^+ \hat{I}_m^- + \hat{I}_n^- \hat{I}_m^+ - 4\hat{I}_n^z \hat{I}_m^z], \quad (2)$$

with the dipolar interaction strength given by $d(\mathbf{r}) = [1 - 3(r_z/r)^2]/4r^3$, and

$$\hat{H}_{E-N} = \sum_{i,n} \gamma_E \gamma_N h_i(\mathbf{R}_i - \mathbf{r}_n) \hat{S}_i^z \hat{I}_n^z, \quad (3)$$

$$h_i(\mathbf{R}) = \frac{8\pi}{3} |\Psi_i(\mathbf{R})|^2 - \int d^3r |\Psi_i(\mathbf{r})|^2 \frac{|\mathbf{r} - \mathbf{R}|^2 - 3[r_z - R_z]^2}{|\mathbf{r} - \mathbf{R}|^5}, \quad (4)$$

written in atomic units; a factor of $\hbar/(4\pi\epsilon_0)$ is implied for the Hamiltonian. The hyperfine interaction, $h_i(\mathbf{r})$, may be approximated by the dipolar interaction, $d(\mathbf{r})$ when \mathbf{r} is far outside the wave function of donor i . Electron spin operators are written as \hat{S} with i or j indices and \mathbf{R}_i position vectors. Nuclear spin operators are written as \hat{I} with n or m indices and \mathbf{r}_n position vectors. The gyromagnetic ratios of the electron and ^{29}Si nuclear spins are $\gamma_E = 1.76 \times 10^{11} (\text{T s})^{-1}$ and $\gamma_N = 5.31 \times 10^7 (\text{T s})^{-1}$ respectively. The wave function of each donor electron, $\Psi_i(\mathbf{r})$, is the Kohn-Luttinger wave function of a phosphorus donor impurity in silicon, as described in Ref. [11]. In addition to the Hamiltonian-governed free evolution, we model spin echo refocusing pulses as ideal spin flips.

To compute the decoherence time of a qubit in Si:P system, we take one of our donor electrons to be the “central” spin, say $i = 0$, and simulate a Hahn spin echo on that donor electron to remove the effects of static noise. Our dominant decoherence is due to flip-flopping bath spins: ^{29}Si -induced and donor-induced SD. We display agreement with experiment for over 5 orders of magnitude in C_N , Fig. 1, maintaining agreement into very sparse densities. This Letter presents procedures we have developed to accomplish this substantial (and very computationally demanding) task.

We previously [4] computed the decoherence for $C_N \gtrsim 1000$ ppm using a cluster expansion technique which works well for dense spin baths. For sparse baths, we use the CCE [14], applicable to both small and large spin baths, with some adaptations. The CCE has a simple and self-evident formulation which we now describe. We define $\mathcal{L}(t) = \rho_{\uparrow\downarrow}(t)/\rho_{\uparrow\downarrow}(0)$, the off-diagonal element of the reduced density matrix of our central spin after performing a spin echo sequence over the duration $t = 2\tau$, a refocusing pulse occurring at time τ . The spin echo figure of merit is the modulus of $\mathcal{L}(t)$. Next, for a given set (cluster) of electron or nuclear bath spins, \mathcal{C} , we define $L_{\mathcal{C}}(t)$ to be the resulting $\mathcal{L}(t)$ when we only include flip-flop terms in our Hamiltonian [Eqs. (1-3)] that involve elements of \mathcal{C} ; all $\hat{S}_i^z \hat{S}_j^z$ interactions are included in our implementation. Then, we recursively define

$$\tilde{L}_{\mathcal{C}}(t) = L_{\mathcal{C}}(t) / \prod_{\mathcal{C}' \subset \mathcal{C}} \tilde{L}_{\mathcal{C}'}(t). \quad (5)$$

By tautology, $\mathcal{L}(t) = \prod_{\mathcal{C}} \tilde{L}_{\mathcal{C}}(t)$, providing a way to break the problem into independent factors coming from each

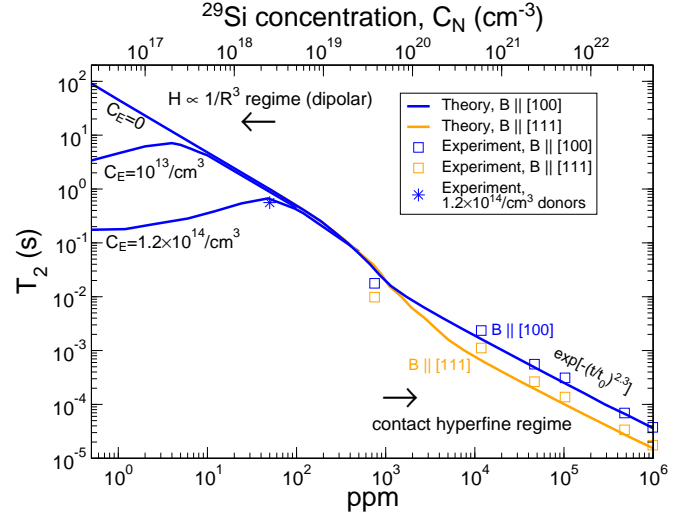


FIG. 1: Decay times (T_2 for the Hahn echo) of Si:P donor electron spins for various C_N . At high C_N , contact hyperfine interactions dominate and T_2 is dependent upon the magnetic field direction relative to the lattice orientation. At low C_N , T_2 is dependent upon C_E , and eventually dominated only by dipolar interactions (which includes dipolar-approximated electronuclear interactions). Experimental results are shown as square symbols, from Ref. 2, and a star symbol, from Ref. 3.

set of bath spins. At short times, the smallest nontrivial clusters dominate the decay; successively larger clusters become significant with increasing evolution time.

These cluster expansions work well by perturbative arguments in the regime where the interactions among the bath spins are weak relative to the interaction with the central spin. Thus, ^{29}Si -induced SD is well-approximated when including only 2-clusters. Donor-induced SD, however, is much more challenging because the interaction strengths among the bath spins and with the central spin are comparable. In fact, we find that if we compute the CCE expansion for different spatial configurations and different initial spin states, the average over these configurations and states can diverge rapidly when we include 4-clusters. We attribute this to the fact that different configurations of a sparse bath, or even different states of a given spatial configuration, can have very different convergence time scales for CCE. However, we find that the CCE is well behaved if we average over spin states within the CCE definitions, that is, $\mathcal{L}(t) = \langle \rho_{\uparrow\downarrow}^J(t) \rangle_J / \langle \rho_{\uparrow\downarrow}^J(0) \rangle_J$, where J represents each spin state and $\rho_{\uparrow\downarrow}^J$ is calculated with J as the initial bath state.

Of course, averaging over all spin states exactly would be prohibitively difficult. We find, however, that it is sufficient to average over spin states in the following self-consistent manner. Choose a spatial configuration and a spin state $|J\rangle = \bigotimes_n |j_n\rangle$ that serves as a template for spin state variants. Let Γ be a set of clusters (e.g., up to a

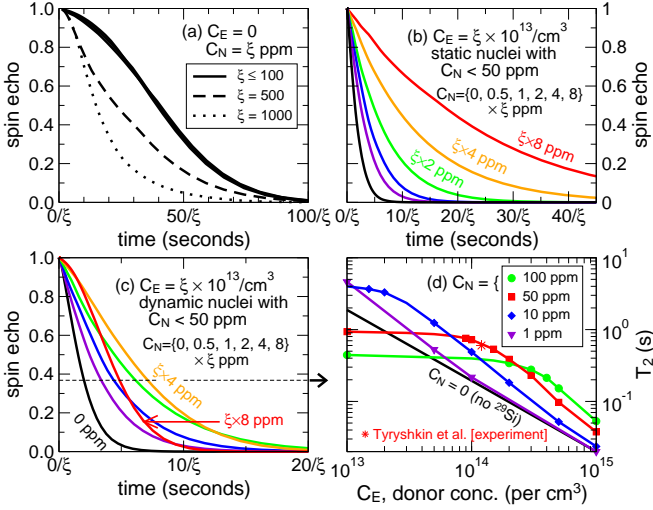


FIG. 2: Spin echo resulting from decoherence induced by various concentrations of (a) ^{29}Si (C_N), (b) background phosphorus donors (C_E), or (c) the combination of both. In (b), static ^{29}Si -induced Overhauser field variations between donors suppresses their decoherence-inducing flip-flops. (a)-(c) use ξ as a scaling parameter for both concentration and inverse time. (d) T_2 , defined as the $1/e$ decay time, for various C_N and C_E . The star symbol indicates an experimentally obtained[3] result at $C_N \approx 50$ ppm and is in good agreement with the corresponding 50 ppm theoretical curve.

certain size) that we include to approximate the solution:

$$\mathcal{L}_\Gamma^J = \prod_{C \in \Gamma} \tilde{L}_C^{K(J,C,\Gamma)}, \quad (6)$$

where $\mathcal{K}(J, C, \Gamma)$ is the set of all spin states that may differ from J only for spins in superclusters of C that are contained in Γ . That is,

$$\mathcal{K}(J, C, \Gamma) = \{J' | \exists C' \in \Gamma, C' \supseteq C, \mathcal{D}(|J\rangle, |J'\rangle) \subseteq C'\}, \quad (7)$$

where $\mathcal{D}(|J\rangle, |J'\rangle)$ is the set of spins whose state differs between $|J\rangle$ and $|J'\rangle$. Then we define

$$\tilde{L}_C^K = \langle L_C^K \rangle_{K \in \mathcal{K}} / \prod_{C'' \subset C} \tilde{L}_{C''}^K, \quad (8)$$

where L_C^J solves the L_C problem for the given spin state J . Importantly, this yields the exact spin state average solution for \mathcal{L}_Γ^J in the limit that Γ includes all clusters (J becomes irrelevant). Furthermore, it may be computed relatively efficiently. With proper bookkeeping, each Hamiltonian (for a given cluster and external spin state) need only be diagonalized once, and each L_C^J need only be computed once and raised to the proper power to be multiplied into the solution.

We use heuristics and cutoffs to determine the Γ set of clusters to include, trying to minimize the set necessary to approximate the solution well. We heuristically favor clusters with strong interactions forming a connected

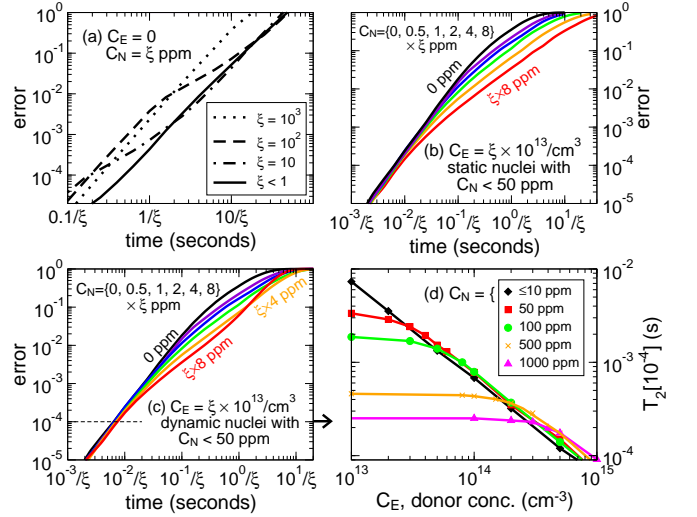


FIG. 3: (a)-(c) correlate with (a)-(c) of Fig. 2, respectively, but plotted in terms of error (one minus the spin echo) on a log-log scale in order to highlight the low-error behavior that is relevant for fault-tolerant quantum computation. (d) Plots $T_2[10^{-4}]$, the time at which the echo error is 10^{-4} , for various C_N and C_E .

graph over the entire cluster and we employ cutoffs in the number of clusters, resonance energies, and distance from the central spin. We compute ensemble average results, such as shown in Figs. 1, 2, and 3, by averaging results of different spatial configurations and J spin state templates for a given set of cutoffs. These cutoffs are adjusted until we obtain consistent, convergent results.

We present, in Figs. 2 and 3, ensemble averaged spin echo results for varied C_E and C_N , both separately and combined. We use ξ as a scaling parameter to illustrate a perfect correspondence between concentrations and inverse time when decay is dominated entirely by $1/r^3$ dipolar interactions ($C_N \lesssim 50$ ppm). These results show behavior ranging from decay dominated by ^{29}Si -induced SD to decay dominated by donor-induced SD. When not dominated by ^{29}Si -induced SD, the presence of ^{29}Si can actually prolong coherence, see Figs. 1 and 2 (d), because Overhauser field variations suppress donor flip-flops; a similar effect is noted [10] with respect to qubit concentration and layout. The initial decay, shown clearly in Fig. 3, behaves differently; in this regime, any beneficial effect of ^{29}Si is fairly insignificant.

Computing or measuring ensemble averages, as shown in Figs. 2 and 3, has limited utility in the scope of quantum computation. It is more informative to compute the full distribution of results that come out of the considerable sample-to-sample variation, which are significant especially for a central spin with dipolar coupling to a dilute bath [15]. Figure 4 addresses this for donor-induced spectral diffusion by showing error distribution information for each spin echo time independently; essentially, this gives a performance guarantee for various

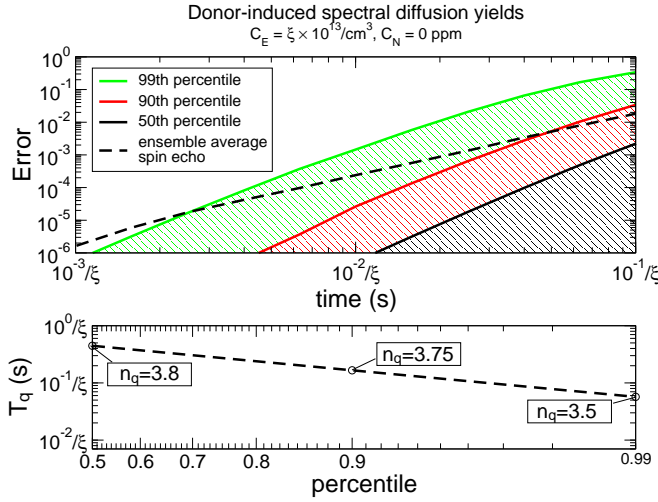


FIG. 4: (top) Maximum error from donor-induced (without ^{29}Si) SD for at least 50%, 90%, or 99% of the central spins due to random spatial variations of the bath spins. (bottom) Corresponding fits of low-error behavior of $1 - \exp[-(t/T_q)^{n_q}] \approx (t/T_q)^{n_q}$ in the 10^{-4} error regime.

fractions of possible donors. At short times, the ensemble average echo decay error is actually dominated by statistical outliers as the top panel of Fig. 4 demonstrates. Used in the bottom panel of Fig. 4, we introduce T_q and n_q as figures of merit that characterize initial decoherence at short times, as appropriate for quantum information considerations. These are obtained by fitting the error to $1 - \exp[-(t/T_q)^{n_q}] \approx (t/T_q)^{n_q}$ in the 10^{-4} error regime (motivated by common fault-tolerance thresholds). These results have direct implications for quantum computer architecture designs and error analysis [16].

Apart from reducing C_E , donor-induced SD may be suppressed by polarizing the background donors thermally at temperatures that are readily achieved in specialized refrigerators [17]. In the low-error limit where donor-induced SD often dominates [Fig. 3], the error is proportional to the number of contributing 2-clusters which must have opposite spin polarization in order to flip-flop; thus, $T_q \propto (p_\uparrow p_\downarrow)^{-n_q}$, where $p_{\uparrow/\downarrow} = \exp(\pm E_Z/k_B T)/2 \cosh(E_Z/k_B T)$ from Boltzmann statistics with E_Z as the electron Zeeman energy splitting corresponding to about 1.3 K per Tesla.

To conclude, we adapted the cluster correlation expansion [14], by retaining all Ising-like interactions and interlacing spin state averaging in a self-consistent manner, to study decoherence induced by a background of dynamical donor electron spins in silicon. We demonstrate that approaching the $T_2 = 2T_1$ limit through isotopic enrichment in Si is impossible in the presence of a finite concentration of unpolarized donors. Unavoidable donor impurities in the background make this limit, where T_1 of one hour has been reported at 1.25 K [18], unattainable, though prospects improve if the electrons may be

thermally polarized. While the presence of some ^{29}Si can actually increase T_2 considerably by suppressing donor-induced decoherence, this effect is fairly insignificant in the short time (low-error) regime important for quantum computation. We introduce T_q and n_q to describe decoherence at short times and discuss the effect of statistical variation of impurity locations on decoherence. Variation in the decoherence of different donors becomes extremely significant in the regime of low impurity concentration, a crucial consideration for designing quantum computer architectures and determining fabrication requirements.

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